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THE PRECISION OF NUCLEAR DECAY RATES†

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It is commonly supposed that radioactive isotopes decay in a strictly exponential way, so that the process can be characterized by a half-life; and that the half-life depends only on the isotope, not being influenced at all by surroundings. Now both of these assumptions are challenged: it is questioned whether the decay is always strictly exponential, and there is evidence to show that in some cases at least the decay may be influenced by the surroundings, or by something else external to the nuclei. The importance of this possibility in trying to establish ages with the use of carbon 14 is obvious; and the question is of first-rate importance for physics generally.

Introduction

Each of the 1600 known radioactive isotopes has a characteristic rate of decay measured in terms of half-life, $t_{1/2}$. This $t_{1/2}$ is defined as the time required for the decay of one-half of the original excited nuclei.

The precision of nuclear decay rates refers to the exactness and constancy of these measured lifetimes. Such precision is a basic assumption of all radiometric dating techniques. In addition this assumption of constant $t_{1/2}$ is stated as fact in nearly every text book which has treated radioactivity since its discovery by Becquerel in 1896.

The high energies involved in nuclear interactions are thought to make nuclear parameters entirely independent of external conditions. However there is growing evidence and awareness that nuclear half-lives are variables rather than constants. Journal editorials[1] and articles[2,3] are mute evidence that nuclear physics remains an experimental science.

The implications of variation of nuclear decay rates in the past and their possible control in the future are great. First, all experimental $t_{1/2}$ measurements must recognize the added parameter of nuclear environment. Much $t_{1/2}$ literature is incomplete because the chemical matrix of the nuclei and the laboratory conditions are not specified. All past and future half-life analysis must take into account variation of results depending on extranuclear conditions.

Second, a re-evaluation of radiometric dating and geochronology is needed. There is strong resistance to this specific challenge because radiometric dating results are much publicized.

Third, the control of the time dimension of radioactivity provides a potential energy source. Short nuclear half-lives could conceivably be lenghtened and long lives telescoped to provide controlled energy release from decaying nuclei. Also the telescoping of long half-lives could rapidly decontaminate radioactive wastes, thus eliminating one of nuclear energy's major drawbacks.

Fourth, in view of the variability of half-life values a study of other physical constants, laws, and assumptions is in order.

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Theory

(1) Half-Life Values: The half-life used to catalog radioactive isotopes may be defined in several ways. In completely random decay events the usual decay equation holds,

$$N = N_0 e^{-\lambda t}. \quad (1)$$

Here N_0 and N are respectively the number of radioactive atoms initially and at a time t. The parameter λ represents the individual decay probability per unit time.

This equation is an approximation since the decay process under perturbation conditions is not random and is not properly described by the Poisson distribution assumed in Equation 1.[4] In the perturbation case λ depends on the nuclear environment, the subject of this paper. Half-life varies inversely with λ,

$$t_{1/2} = \frac{1n2}{\lambda}. \quad (2)$$

The nuclear half-life also appears in the Heisenberg uncertainty principle relating energy and time,

$$t_{1/2} \geq \frac{h\ 1n2}{\Gamma 2\pi} \quad (3)$$

The energy uncertainty Γ is the width of the excited nuclear state before decay. The time uncertainty is just the half-life, related inversely to the linewidth Γ through Planck's constant h. Note that as uncertainty in nuclear decay energy increases due to broadening by extranuclear interactions, the half-life necessarily decreases.

The inequality sign in Equation 3 is needed when the nucleus is perturbed by its environment, the usual case. Thus the equal sign is invalid along with the long-standing assumption that nuclear events are independent of all external considerations. The equal sign only applies in the case of a free isolated nucleus.

Either definition above shows that $t_{1/2}$ cannot be calculated from theory or from other data such as decay energy. It must be measured experimentally and cannot be known exactly.

The neutron is a good illustration of the profound mystery surrounding nuclear decay. Free neutrons have a $t_{1/2}$ of about 12 minutes. However neutrons bound within a stable atomic nucleus become entirely secure and unradioactive. Thus the lifetime of bound neutrons is entirely unrelated to that of free neutrons.

Whether or not they maintain a neutron nature within the nucleus is as unknown as why they decay in the free state. Thus in this discussion of radioactive half-life it is well to keep in mind the very limited understanding of decay events.

Published half-life values have a range of 50 orders of magnitude. Extremes include ^{8}Be (10^{-23} seconds) and ^{144}Nd (10^{+23} seconds). Few other physical properties are measurable over such a large range of values. In view of difficulties (impurities, low counting rate, changes in counting geometry between samples, loss of volatile material) in measuring the longer half-lives, it might be more realistic to say all isotopes are radioactive to some extent, and those with $t_{1/2}$ greater than 10^{10} years are stable. The division between stable and unstable nuclei is arbitrary. Measurement uncertainty increases for both long and short isotopes; ±2% accuracy is typically and optimistically claimed.[5]

(2) First Order Interation: The energies involved in nuclear reactions are usually much larger than the energies of chemical binding. Nevertheless interactions do result from energy coupling between the nucleus and surrounding electrons.

The effect is seen as a perturbation of the free nuclear-electron Hamiltonian, the function which specifies the energy state of the nucleus. This perturbation produces shifts and splittings of the nuclear energy levels which secondarily perturbs all nuclear parameters including decay rate.

Consider the electrostatic interaction between a nucleus and the electron charge with which it interacts. Let ρ represent a uniform s-electron density. These inner electrons have a finite probability of actually overlapping the nuclear site. Quantum mechanically the s-electron wave functions show penetration of the nucleus for a fraction of the time.

For a simple model assume the nucleus is a uniformly charged sphere of radius R. The shift of the decay energy as a result of the electron-nucleus coupling is then shown by comparing the electrostatic interaction for a hypothetical point nucleus and one of actual radius R.

For a point nucleus of charge Ze where Z is the atomic number and e is the unit electron charge, the electrostatic potential V at a distance r from the origin in electrostatic units is

$$V_{point} = \frac{Ze}{r}. \tag{4}$$

For a finite nuclear volume the potential is calculated from the definition

$$V_{finite} = \int_r \frac{dq}{r}. \tag{5}$$

where dq is the nuclear charge increment. The result for the uniformly charged sphere is

$$\begin{aligned} V_{finite} &= \frac{Ze}{R}\left[\frac{3}{2} - \frac{r^2}{2R^2}\right] \qquad r \leqslant R \\ &= \frac{Ze}{r} \qquad\qquad\qquad\quad r \geqslant R \end{aligned} \tag{6}$$

The decay energy shift ΔE is given by an integral over the spherically symmetric region of nucleus-electron overlap,

$$\begin{aligned} \Delta E &= \int_0^R \rho(V_{finite} - V_{point}) 4\pi r^2\, dr \\ &= \frac{4\pi\rho Ze}{R}\int_0^R \left[\frac{3}{2} - \frac{r^2}{2R^2} - \frac{R}{r}\right] r^2\, dr \\ &= \frac{-2\pi Ze\rho R^2}{5} \end{aligned} \tag{7}$$

This result shows that nuclear decay is in general not independent of its electron environment. Decay depends on the energy state of the entire atom rather than on the nuclear state alone.

Experimental confirmation of the perturbation thus far has been mainly limited[6] to atoms with small Z such as ^{7}Be. The reasoning is that s-electrons in this case are also valence electrons, and their density can be easily changed by external effects.

This emphasis on light nuclei is unnecessary, however, since s-electrons spend a fraction of time further from the nucleus than either p or d electrons. Valence effects on outer electrons of high Z atoms will still perturb the nucleus by screening the s-electrons. This phenomenon is presently seen with Mössbauer spectroscopy.[7]

A variety of experimentors have succeeded in changing nuclear decay rates several percent by changing the inner electron density of atoms. These techniques include:

Chemical Effects
Bonding and valence effects
Stress in molecular layers
Physical Effects
Applied electric and magnetic fields
Applied pressure
Magnetic and electric ordering transitions
Superconducting transition
Temperature extremes

(3) Higher Order Interactions: Other perturbations of the nucleus occur besides electrostatic coupling. Nuclei in energy states with nuclear spin other than 0 or ½ do not fit the spherical model. They are nonspherical and their resulting nuclear quadrupole moment $\overline{Q}$ interacts with any electric field gradient $\nabla\overline{E}$ present at the nuclear site. This field gradient may arise either from charges within the sample itself or by external application. The Hamiltonian H_Q in this case is

$$H_Q = \overline{Q} \cdot \nabla\overline{E}\,. \tag{8}$$

An applied or naturally occuring internal magnetic field $\overline{H}$ couples with the nuclear magnetic moment $\overline{\mu}$, another nuclear parameter. This magnetic interaction can be written as

$$H_M = -\overline{\mu} \cdot \overline{H}\,. \tag{9}$$

These quadrupole and magnetic effects shift and split the nuclear energy levels. Just as the electrostatic coupling has been shown to control the nuclear half-life, so these higher order interactions affect the decay rate in ways not exactly known.

(4) Radiation Interactions: Radioactivity should be influenced by incident cosmic particles, electromagnetic radiation, and the disintegration of neighboring nuclei within the sample. A quantitative evaluation of cosmic ray effects involves a comparison of nuclear cross sections with the present and past cosmic ray flux.

However each of these quantities is uncertain. There is a particular shortage of reliable data on nuclear absorption cross sections, one reason why it is impossible to predict closely the efficiency of nuclear power reactors.

One example of cosmic ray effects may be seen in the incredible differences in age shown by returned lunar samples. Since the age of the moon is thought to be of the same order as that of the earth, scientists believed that lunar samples would very accurately correlate with terrestrial dating results.

Instead the radiometric lunar ages vary greatly between 2 million and 28 billion years. Neither value is reliable; and

it may be that the cosmic ray and solar wind particles which are incident on the moon unimpeded by a strong magnetic field or an atmosphere have grossly affected the decay rates of the products being measured.

A special radiation technique which has successfully controlled decay rates involves the construction of a resonant barrier around the nuclei of interest. Nuclei in the excited state are surrounded by identical nuclei in the ground state.

Some of the de-excitation gamma rays from the center region may be absorbed by the barrier and re-radiated back to the original central nuclei. The resonant process continues and has been used to lengthen the radioactive life of an original group of ^{57}Fe nuclei by 3%.[8]

This technique represents a mechanical control of the decay products rather than an actual control of the decay process. Only the nuclear energy levels that show resonance fluorescence qualify for this approach.[9]

(5) Special Effects: Gamow[10] has speculated that all fundamental constants of nature may change with time. Hence the electric charge, speed of light, gravitational constant, and nuclear half-lives are all included as changing parameters that characterize the state of the universe.

Tests of this concept have been negative and thus limit any changes to an infinitesimal scale. An additional argument against this constant drift appears in Psalm 89:2; God's faithfulness is established in the stability of the physical universe.

Relativistic motion of a radioactive source lengthens the nuclear half-life by dilation of time with respect to a stationary observer. For 15 years this phenomenon has been measured for radioactive muons which are formed in the upper atmosphere by cosmic-ray-air molecule collisions.

Muons at rest have a $t_{1/2}$ of about 2 microseconds. When they are traveling at 99% of the speed of light, the $t_{1/2}$ increases 16 times to 32 microseconds. This special case which shows a relative aspect of time actually produces the greatest $t_{1/2}$ variation yet measured.

Data

Early efforts to disprove the assumption of unalterable radioactivity include Kelvin's objections to spontaneous decay in 1905 and Segre's theoretical perturbation predictions in 1947. Today experiments with a variety of nuclei show the control of half-life (Table 1). The changes reported thus far in half-life range from very small to 5.7% for the 73 electron volt metastable state of ^{235}U.

The majority of nuclei examined have been characterized by internal conversion or electron capture, decay types which are especially sensitive to the chemical environment. Notice that the percent variations shown in Table 1 refer to nuclear half-life values, not to radiometric dating ages which are much less reliable.

The list of experimentally perturbed half-lives is growing rapidly. Actually many other isotopes with very short half-lives could be included in Table 1, as for example the 100 different detected Mössbauer Effect gamma transitions. However the longer lived isotopes are of more dramatic interest.

Figures 1 and 2 are presented to show the problems involved in a typical literature summary of $t_{1/2}$ data for ^{24}Na and ^{134}Cs isotopes.[11] For these isotopes authors regularly publish $t_{1/2}$ uncertainties that lie entirely outside of the error bars of each other. Either the half-life is varying between samples, the authors do not understand statistical uncertainty, or both are true. It is evident that much nuclear lifetime data are untrustworthy; separate measurements simply are not the same.

TABLE 1

Radioactive isotopes showing experimental $t_{1/2}$ variation (References 1, 3, 13). Abbreviations: ns (nanosecond, 10^{-9} second), s (second), m (minute), h (hour), d (day), y (year), EC (electron capture), β- (electron emission), β^+ (positron emission), γ (gamma radiation). (Note: The last item, the uranium, is in a metastable state.)

Isotope	Average Half-Life	Half-Life Percent Variation	Decay Mode
^{7}Be	53.5 d	0.18	EC
^{14}C	5730 y	small	β^-
^{24}Na	15 h	0.5	β^-
^{57}Fe	100 ns	3	γ
^{60}Co	5.26 y	small	β^-
^{64}Cu	12.8h	1.7	EC, β^-, β^+
^{85}Sr	65.2 d	0.005	EC
^{89}Zr	78.4 h	0.08	EC, β^+
^{90}Nb	24 s	3.7	γ
^{96}Nb	23.5 h	3.6	EC
^{99}Tc	6 h	0.31	γ
^{119}Sn	18 ns	small	γ
^{121}I	2.12 h	small	EC, β^+
^{125}Te	58 d	0.026	γ
^{131}I	8 d	0.3	β^-
^{134}Cs	3 h	small	γ
^{137}Cs	30 y	small	β^-
^{169}Tm	4 ns	small	γ
^{193}Pt	9.7 ns	4.0 +2	γ
^{235}U	26.1 m	5.7	EC

The generality of this problem and the tremendous economic loss due to worthless data are seen in the current catalog of isotope half-lives.[12] Listed are 57 isotopes with more than 10% variation in measured half-life between researchers, and 35 isotopes with more than a 100% difference in published $t_{1/2}$ values.

Figures 1 and 2 in addition show a tracking phenomenon. Tracking is the tendancy of reported measurements to cluster about an incorrect value. Researchers are reluctant to report findings too far different from previous results in their published findings, or defective measurements or assumptions are repeated. Another large-scale example of this clustering effect is seen in the 4.5 billion year age assumption for the earth.

Conclusion

Radioactive nuclei lifetimes have been controlled by a variety of methods. There remains a strong inertial tendancy from science text publishers to ignore this new time dimension of $t_{1/2}$. Certainly a new model of the atom is needed in which nuclear reactions involve the entire atom. Half-life must be considered as a stability index, a complex variable with extranuclear dependence.

Presently available nuclear data are very unclear as to possible influence by variable $t_{1/2}$ values. Accuracy of the numerical data reported in the scientific literature cannot

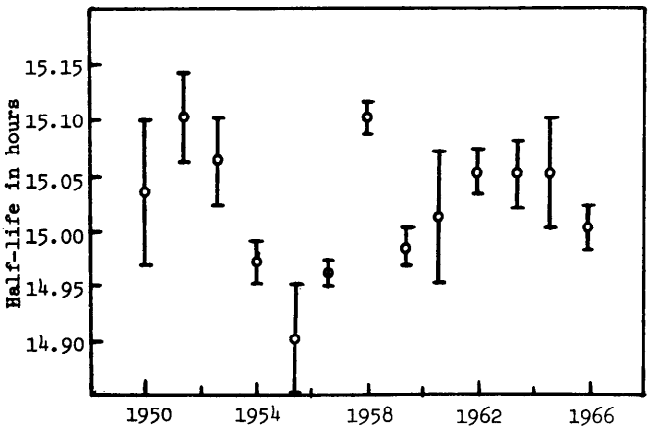

Figure 1. Published values of the half-life of ^{24}Na over a recent period of 20 years.

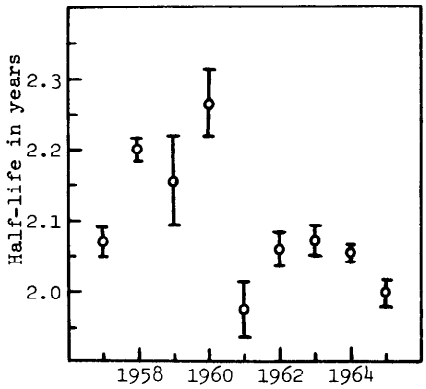

Figure 2. Published values of the half-life of ^{134}Cs over a recent period of 10 years.

be determined from the evidence presented. Concurrent error ranges for single isotopes are not the same. Many experiments are underway to increase the list of perturbed lifetimes and to increase the magnitude of variation. It will be of interest to watch this area of $t_{1/2}$ research expand, and watch researchers become more cautious as the implications for geochronology, energy, and nuclear theory are realized.

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